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Temporal trends of legacy organochlorines in eggs of Canadian Arctic seabirds monitored over four decades

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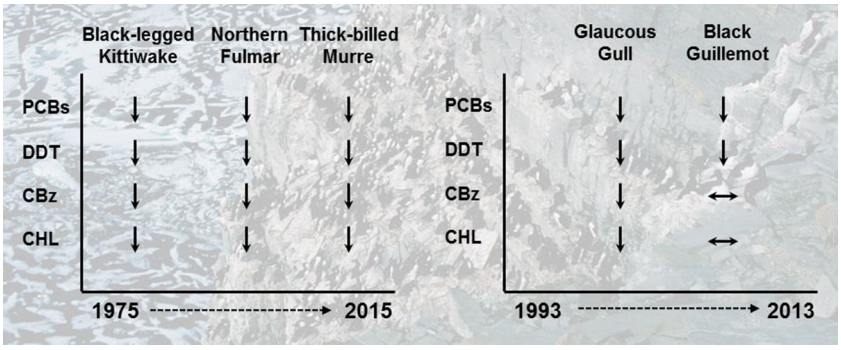
HIGHLIGHTS

- The major organochlorines found in seabird eggs were Σ_{35}PCB, ΣDDT, ΣCBz and ΣCHL.
- Most legacy organochlorines declined since 1975 in Canadian Arctic seabird eggs.
- Most of the declines occurred during the 1970s to 1990s.
- β-HCH continues to increase in eggs of most Canadian Arctic seabird species.
- Glaucous gulls generally had the highest organochlorine concentrations.

GRAPHICAL ABSTRACT



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ABSTRACT

We compared temporal trends of legacy organochlorine pesticides and PCBs in eggs of five seabird species breeding at Prince Leopold Island in the Canadian high Arctic. Concentrations of most of the major organochlorine groups/compounds have either declined (e.g. Σ_{35}PCB, ΣDDT, ΣCBz, ΣCHL, octachlorostyrene) or shown no consistent directional change (e.g. heptachlor epoxide) since 1975 in eggs of thick-billed murres (*Uria lomvia*), northern fulmars (*Fulmarus glacialis*) and black-legged kittiwakes (*Rissa tridactyla*). Aside from β-HCH, which increased in most species, the major organochlorine compounds either declined or showed no trend between 1993 and 2013 in eggs of five seabird species (thick-billed murre, northern fulmar, black-legged kittiwake, black guillemot *Cepphus grylle*, glaucous gull *Larus hyperboreus*). Most of the declines occurred during the 1970s to 1990s followed by little change during the 2000s. Glaucous gull eggs had the highest concentrations of almost all organochlorines in the five years compared (1993, 1998, 2003/04, 2008, 2013), and murre eggs generally had among the lowest concentrations. The primary organochlorines found in eggs of all five species were Σ_{35}PCB, ΣDDT (mainly *p,p′*-DDE), ΣCBz (mainly hexachlorobenzene) and ΣCHL (mainly oxychlordane) although proportions varied by species and year. The major PCB congeners found in eggs of all five species were CB-153, -138, -118 and -180. The penta-, hexa- and heptachlorobiphenyl homologs comprised the largest proportion of Σ_{35}PCB in all five species. Although levels of most legacy organochlorines have declined since 1975, the potential for climate change to alter chemical transport pathways as well as exposure pathways in the biotic environment could affect temporal trends. Therefore, it is important to continue to monitor these legacy contaminants in order to determine how these changes will affect the temporal trends observed to date.

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1. Introduction

The presence of persistent organic pollutants (POPs) has been monitored in the Arctic environment since the early 1970s (AMAP, 2016; NCP, 2013; Rigét et al., 2010). Those POPs which have been banned or regulated under international conventions, such as the Stockholm Convention on POPs (http://chm.pops.int) and the United Nations Economic Commission for Europe (UN ECE) Convention on Long-range Transboundary Air Pollution (LRTAP) Protocol on POPs (http://www.unece.org/env/lrtap/pops_h1.html), are often referred to as "legacy" POPs because present day contamination or re-emissions are primarily related to a "legacy" of past releases (Rigét et al., 2010). As a result of national and international efforts to reduce emissions, legacy POPs, such as organochlorine pesticides (e.g. DDT) and PCBs, have generally declined in the Arctic environment (AMAP, 2016; NCP, 2013). Declining trends of legacy POPs observed in Arctic biota are consistent with decreasing trends in Arctic air which reflect the historic decreases in emissions (AMAP, 2016; Rigét et al., 2010). However, there is renewed interest in legacy POPs due to the potential for climate change to alter chemical transport pathways as well as exposure pathways in the biotic environment (Borgå et al., 2010; Ma et al., 2011) which could affect temporal trends.

The statistical power of a time-series to detect a trend increases with the number of years of sampling data (Fryer and Nicholson, 1993). For most Arctic time-series, the statistical power to detect a trend of the magnitude typically observed for Arctic biota is rather low (AMAP, 2016; Rigét et al., 2010). Contaminant time-series for seabird eggs sampled from the Canadian high Arctic have provided the highest power and among the lowest detectable trends for Arctic biota (AMAP, 2016). At the time of egg formation, lipophilic organochlorine compounds are transferred along with lipids to the eggs thus reflecting the relative contaminant burden of the various POPs in the female at the time of laying (Braune and Norstrom, 1989; Verboven et al., 2009). Therefore, eggs are an ideal and relatively non-intrusive sampling medium.

Eggs of thick-billed murres (*Uria lomvia*), northern fulmars (*Fulmarus glacialis*) and black-legged kittiwakes (*Rissa tridactyla*) have been monitored for environmental contaminants at Prince Leopold Island, Nunavut, in the Canadian high Arctic since 1975 (Braune, 2007). As of 1993, eggs of black guillemots (*Cepphus grylle*) and glaucous gulls (*Larus hyperboreus*) have also been monitored for contaminants at that location. Four of those species, the thick-billed murre (a.k.a. Brünnich's guillemot), northern fulmar, black-legged kittiwake and black guillemot, feed in the marine environment (Bradstreet, 1980; Butler and Buckley, 2002; Hatch et al., 2009; Mallory et al., 2012; Matley et al., 2012; Provencher et al., 2012), while the glaucous gull feeds in both marine and terrestrial environments (Weiser and Gilchrist, 2012), although the gulls at Prince Leopold Island probably feed principally on marine prey during the breeding season (Matley et al., 2012; Nettleship et al., 1990). Despite the fact that they all have a marine-based diet during the breeding season, their trophic positions vary considerably (Braune et al., 2016), thus influencing their exposure to contaminants.

The objectives of this study were to: (i) compare concentrations of legacy organochlorine contaminants as well as PCB congener patterns in eggs of five seabird species breeding in the Canadian high Arctic, (ii) examine temporal trends of legacy organochlorines in eggs of those same five seabird species to determine if the trends among species are similar over a given time period, and (iii) extend the time-series data for organochlorines in eggs of three of those seabird species previously reported by Braune (2007).

2. Materials and methods

2.1. Sample collection and preparation

During 1975–2015, eggs of thick-billed murres, northern fulmars, black-legged kittiwakes, black guillemots, and glaucous gulls were collected from the Prince Leopold Island Migratory Bird Sanctuary (74°02′N, 90°05′W) in Lancaster Sound, Nunavut, Canada (Fig. S1). Murre and fulmar eggs were sampled in 1975, 1976, 1977, 1987, 1988 (murre eggs only), 1993, 1998, 2003, annually from 2005 to 2014, and in 2015 (fulmar eggs only). Kittiwake eggs were sampled in 1975, 1976, 1987, 1993, 1998, 2003, 2008, and 2013; black guillemot eggs in 1993, 1998, 2004, 2008, and 2013; and glaucous gull eggs in 1993, 1998, 2003, 2008, and 2013. Thick-billed murres and northern fulmars lay a single egg, whereas black-legged kittiwakes and glaucous gulls may lay up to three eggs (Gaston et al., 2005). Black guillemots generally lay one or two eggs (Butler and Buckley, 2002). Eggs were sampled randomly on the basis of one egg per nest as soon after laying as was possible. Eggs were collected by hand or using a small cup attached to the end of an extension pole. All eggs were taken under appropriate annual research and collection permits.

Eggs were kept cool in the field and shipped to the National Wildlife Research Centre (NWRC), Ottawa, Ontario, for processing and chemical analyses. Egg contents were homogenized and stored frozen (−40 °C) in acetone-hexane rinsed glass vials for subsequent organochlorine analysis. The validity of this storage method has been previously discussed (see Braune, 2007).

Three to 15 eggs per species per year were analyzed (see Tables S1–S5 for sample sizes). Egg homogenates from 1975 to 2005 were analyzed for organochlorines and $\delta^{15}N$ as pooled (composite) samples with each pool comprising equal aliquots of three individual eggs, with the exception of glaucous gull eggs sampled in 1993, which were analyzed as pools of two eggs each. Eggs sampled during 2006–2015 were also analyzed for organochlorines as pooled samples but were individually analyzed for $\delta^{15}N$.

2.2. Organochlorine analysis

Organochlorine analyses were carried out at NWRC. Archived samples collected prior to 1998 were retrieved from the National Wildlife Specimen Bank at NWRC and analyzed retrospectively in 1998–99 in order to standardize pooling and analytical protocols which varied over earlier years of sampling. Archived eggs of black guillemots and glaucous gulls collected in 1993 were re-analyzed in 2006 for the same reasons. Samples collected from 1998 to 2015 were analyzed within six months of collection. Pooled egg homogenates were analyzed for organochlorines (OCs) including chlorobenzenes (ΣCBz = 1,2,4,5-tetrachlorobenzene, 1,2,3,4-tetrachlorobenzene, pentachlorobenzene and hexachlorobenzene), hexachlorocyclohexanes (ΣHCH = α-, β- and γ-hexachlorocyclohexane), heptachlor epoxide (HE), chlordane-related compounds (ΣCHL = oxychlordane, *trans*-chlordane, *cis*-chlordane, *trans*-nonachlor and *cis*-nonachlor), DDT and its metabolites (ΣDDT = *p,p′*-DDE, *p,p′*-DDD and *p,p′*-DDT), octachlorostyrene (OCS), mirex (ΣMirex = photomirex and mirex), dieldrin and PCB congeners (ΣPCB). ΣPCB was standardized to 35 congeners identified according to IUPAC numbers (Ballschmiter et al., 1992): 18, 17 (or 18/17), 31/28, 33 (or 33/20), 52, 49, 44, 74, 70 (or 70/76), 95, 101 (or 101/90), 99, 87, 110, 151, 149, 118, 153, 105, 158, 138, 187, 183, 128 (or 128/167), 177, 171, 156, 180, 170 (or 170/190), 199, 208, 195, 194, 206. Congeners separated by a slash co-eluted during the chromatography process and were therefore reported together.

Samples were analyzed for organochlorines by capillary gas chromatograph coupled with a mass selective detector (GC/MSD) operated in selected ion monitoring electron impact mode and lipids were determined by gravimetric methods. Chemical extraction and cleanup of PCBs and organochlorine pesticides in samples from 1975 to 2014 followed the procedures of Lazar et al. (1992). Tissue homogenates were ground with anhydrous sodium sulfate, spiked with labeled ^{13}C-OC/PCB internal standards and extracted with dichloromethane:hexane (50:50% v/v). Sample clean-up was performed by gel permeation chromatography followed by activated Florisil® column chromatography. Samples analyzed in 2015 were homogenized with diatomaceous

earth instead of anhydrous sodium sulfate, extracted with the aid of accelerated solvent extraction (ASE) instead of gravimetrically using dichloromethane:hexane, and solid phase extraction (SPE) replaced the Florisil® column chromatography in the sample clean-up. Labeled ^{13}C-OC/PCB internal standards included chlorobenzenes and six PCB congeners for the 1975–2008 samples and a comprehensive suite of internal standards (chlorobenzenes, HCHs, octachlorostyrene, chlordane-related compounds, dieldrin, DDT metabolites, mirex, PCB congeners) for the 2009–2015 samples. Duplicate extractions, duplicate injections, method blanks, in-house reference materials (HERG QA Reference Egg Pools and/or DCCO QA Reference Egg Pools) for 1975–2013 samples, and certified reference material (NIST1947 Lake Michigan Fish Tissue) for 2009–2015 samples were run for quality control.

All residue concentrations were corrected for internal standard recoveries from 2009 on. Prior to 2009, typical internal standard recoveries varied between 75% and 95% for most PCBs and organochlorines, and over 60% for the highly volatile compounds (i.e. chlorobenzenes, HCHs). As we had limited labeled internal standards prior to 2009, most residues were not corrected for internal standard recoveries except for the highly volatile compounds (i.e. chlorobenzenes, HCHs) which were recovery-corrected based on internal standard recoveries for the chlorobenzenes. The nominal detection limit was 0.1 ng g^{-1} wet weight (ww) for the 1975–2013 samples and varied by analyte for the 2014–2015 samples as indicated in Tables S1-S5.

2.3. Stable nitrogen isotope analysis

Relative trophic position was estimated based on stable nitrogen isotope ratios (^{15}N/^{14}N, expressed as δ^{15}N). Stable nitrogen isotope analyses were performed on one-mg subsamples of freeze-dried, lipid-extracted egg homogenates according to procedures described in Braune et al. (2015a).

2.4. Data treatment

As samples collected from 2006 to 2015 were individually analyzed for δ^{15}N, those data were averaged for groups of the same three eggs that were pooled for the OC analyses to facilitate statistical comparison with related organochlorine data. Non-detect values were set to one half the detection limit for purposes of statistical analyses but were set to zero for calculation of the sums of major organochlorine groups (i.e. Σ_{35}PCB, ΣDDT, ΣCBz, ΣCHL, ΣHCH, ΣMirex) and for calculation of the annual means which appear in Tables S1-S5. To determine which of the major organochlorine groups/compounds dominated the organochlorine profile, wet-weight concentrations were used to calculate the percent contribution of the major organochlorine groups/compounds to the organochlorine total. Total organochlorine concentrations (ΣOC) were calculated as the sum of ΣCBz + ΣHCH + OCS + HE + ΣCHL + ΣDDT + dieldrin + ΣMirex + Σ_{35}PCB.

Interpretation of contaminant temporal trends in biota may be confounded if populations vary their diet over trophic levels through time (Hebert et al., 1997, 2000). The ratio of ^{15}N/^{14}N (δ^{15}N) measured in eggs reflect the diet of the female prior to, or during, egg-laying (Hebert et al., 1999; Hobson, 1995) and can be used as an index of trophic position (Hebert et al., 1999; Hobson and Welch, 1992; Hobson et al., 1994). As a previous study in Lancaster Sound suggested that shifts in baseline δ^{15}N over the last three decades were minor in comparison to potential isotopic changes associated with changes in seabird diets (Moody et al., 2012), we included δ^{15}N in our statistical analyses to account for any shifts in seabird diet.

All statistical tests were performed using Statistica for Windows Version 7.0 (StatSoft Inc., Tulsa, OK) with a significance level of $p < 0.05$. Only those compounds for which >75% of the samples had detectable concentrations for a given species were statistically analyzed. Changes in percent contributions of individual compounds, isomers, metabolites and PCB homologs to chemical group totals over time were analyzed using the Spearman Rank Correlation. Temporal changes in organochlorine concentrations for each species were examined using generalized linear model (GLM) analyses. The three longest-running data sets (i.e. kittiwakes 1975–2013, murres 1975–2014, fulmars 1975–2015) were first analyzed by a series of GLMs, and we used Akaike's Information Criterion adjusted for small sample sizes (AICc; Burnham and Anderson, 2002) to evaluate whether linear or second-order polynomial regressions better fit the temporal changes in concentrations for each species, and to compare fit of the various models generated with year, year^2, % lipid and δ^{15}N as independent factors. Only those models where the difference between model fit was small (ΔAICc ≤ 2) are presented in Tables 1–3 as candidate models considered to have strong support (Burnham and Anderson, 2002) followed by the next best-fitted model. For comparative purposes, the three longer time series were then reduced to the same five years (1993, 1998, 2003/04, 2008, 2013) for which data were available for black guillemots and glaucous gulls, and those five data sets analyzed using AICc as described above (Tables S6–S10). We ran 825 models for the five species sampled during the same years, and an additional 495 models for the three species sampled across all years, for a total of 1320 models. Concentration data were

Table 1
Akaike's Information Criterion (ΔAICc, corrected for small sample sizes) values for representative models explaining variation in organochlorine concentration in eggs of thick-billed murres breeding at Prince Leopold Island, NU, 1975–2014. Models compared linear and quadratic regressions to describe trends in organochlorine concentrations with year, year^2, %lipid and δ^{15}N as independent factors; model weights are presented (w_i^c) as is the R^2 from each GLM. We present only where the difference between model fit was small (ΔAICc < 2), followed by the next best-fitted model.

Compound[a]	Trend[b]	Model	RSS	*K*	*ΔAICc*	w_i^c	R^2
ΣCBz	↓	δ^{15}N, year, year^2	0.031894	6	0.00	0.72	0.71
		δ^{15}N, %lipid, year, year^2	0.031746	6	1.97	0.27	0.71
		year, year^2	0.037093	4	9.80	0.01	0.67
ΣHCH	↔	%lipid	0.004337	3	0.00	0.40	0.05
		%lipid, δ^{15}N	0.004334	4	2.16	0.16	0.04
α-HCH	↓	year, year^2	0.000545	4	0.00	0.56	0.64
		%lipid, year, year^2	0.000544	5	2.13	0.19	0.63
β-HCH	↑	%lipid, year, year^2	0.002056	5	0.00	0.70	0.32
		δ^{15}N, %lipid, year, year^2	0.002056	6	2.34	0.22	0.38
OCS	↓	year, year^2	0.000012	4	0.00	0.52	0.28
		δ^{15}N, year, year^2	0.000012	5	1.00	0.31	0.27
		%lipid, year, year^2	0.000012	5	2.90	0.12	0.28
HE	↔	δ^{15}N, year, year^2	0.000145	5	0.00	0.23	0.12
		δ^{15}N, year, year^2, %lipid	0.000141	6	0.10	0.21	0.13
		δ^{15}N, year^2	0.000150	4	0.43	0.18	0.10
		δ^{15}N, year, %lipid	0.000146	5	0.55	0.17	0.11
		δ^{15}N	0.000157	3	1.87	0.09	0.07
		δ^{15}N, %lipid	0.000154	4	2.54	0.06	0.08
ΣCHL	↓	δ^{15}N, year, year^2, %lipid	0.002225	6	0.00	0.30	0.30
		δ^{15}N, %lipid, year^2	0.002305	5	0.49	0.23	0.29
		δ^{15}N, %lipid, year	0.002307	5	0.56	0.23	0.29
		δ^{15}N, year, year^2	0.002374	5	2.34	0.07	0.27
ΣDDT	↓	year, year^2	0.093242	4	0.00	0.38	0.56
		year	0.097776	3	1.58	0.17	0.54
		δ^{15}N, year, year^2	0.093242	5	2.28	0.12	0.55
Dieldrin	↓	δ^{15}N, year, year^2	0.0014730	5	0.00	0.49	0.24
		δ^{15}N, %lipid, year, year^2	0.0014309	6	0.47	0.39	0.25
		δ^{15}N, year	0.001607	4	4.69	0.04	0.18
ΣMirex	↔	%lipid, year	0.000166	4	0.00	0.26	0.08
		%lipid, year, year^2	0.000162	5	0.43	0.22	0.09
		%lipid	0.000173	3	1.09	0.15	0.05
		year, %lipid, δ^{15}N	0.000166	5	2.28	0.08	0.07
Σ_{35}PCB	↓	%lipid, year, year^2	0.081472	5	0.00	0.65	0.73
		δ^{15}N, %lipid, year, year^2	0.081387	6	2.26	0.21	0.73

[a] Chlorobenzenes (ΣCBz = 1,2,4,5-tetrachlorobenzene, 1,2,3,4-tetrachlorobenzene, pentachlorobenzene and hexachlorobenzene (HCB)), hexachlorocyclohexanes (ΣHCH = α-, β- and γ-hexachlorocyclohexane), octachlorostyrene (OCS), heptachlor epoxide (HE), chlordane-related compounds (ΣCHL = oxychlordane, *trans*-chlordane, *cis*-chlordane, *trans*-nonachlor and *cis*-nonachlor), DDT and its metabolites (ΣDDT = *p,p′*-DDE, *p,p′*-DDD and *p,p′*-DDT), dieldrin, ΣMirex = photomirex and mirex, and PCB congeners (Σ_{35}PCB).

[b] Trend direction was determined by backward stepwise regression analysis with year, % lipid and δ^{15}N as regressors. Non-significant ($p > 0.05$) time trends are indicated as "↔".

Table 2
Akaike's Information Criterion (ΔAICc, corrected for small sample sizes) values for representative models explaining variation in organochlorine concentration in eggs of northern fulmars breeding at Prince Leopold Island, NU, 1975–2015. Models compared linear and quadratic regressions to describe trends in organochlorine concentrations with year, year^2, %lipid and δ^{15}N as independent factors; model weights are presented (w_i^c) as is the R^2 from each GLM. We present only where the difference between model fit was small (ΔAICc < 2), followed by the next best-fitted model.

Compound[a]	Trend[b]	Model	RSS	*K*	*ΔAICc*	w_i^c	R^2
ΣCBz	↓	%lipid, year, year^2	0.011699	5	0.00	0.58	0.74
		δ^{15}N, %lipid, year, year^2	0.011466	6	0.66	0.41	0.74
		%lipid, year	0.0137710	4	13.76	0.0	0.70
ΣHCH	↔	%lipid, δ^{15}N	0.000276	4	0.00	0.35	0.13
		%lipid, δ^{15}N, year, year^2	0.000266	6	1.53	0.16	0.14
		δ^{15}N	0.000290	3	1.90	0.12	0.10
		%lipid, δ^{15}N, year	0.000275	5	1.97	0.13	0.12
		δ^{15}N, year	0.000289	4	3.82	0.05	0.08
α-HCH	↓	%lipid, year	0.000022	4	0.00	0.56	0.71
		δ^{15}N, %lipid, year	0.000022	5	2.27	0.18	0.71
β-HCH	↑	δ^{15}N, %lipid, year, year^2	0.000164	6	0.00	0.38	0.38
		δ^{15}N, year, year^2	0.000169	5	0.17	0.35	0.37
		δ^{15}N, year^2	0.000180	4	3.13	0.13	0.33
OCS	↓	%lipid, year	0.000005	4	0.00	0.56	0.49
		%lipid, δ^{15}N, year	0.000005	5	2.27	0.24	0.49
HE	↔	%lipid, δ^{15}N, year, year^2	0.000237	6	0.00	0.77	0.38
		%lipid, year, year^2	0.000290	5	2.46	0.22	0.35
ΣCHL	↓	%lipid, year^2	0.045997	4	0.00	0.27	0.35
		%lipid, year	0.046016	4	0.03	0.26	0.35
		%lipid, year, year^2	0.045125	5	0.68	0.19	0.35
		%lipid, δ^{15}N, year^2	0.045834	5	1.97	0.10	0.34
		%lipid, δ^{15}N, year	0.045852	5	2.00	0.10	0.34
ΣDDT	↓	%lipid, year, year^2	0.916386	5	0.00	0.63	0.73
		δ^{15}N, %lipid, year, year^2	0.903653	6	1.16	0.35	0.76
		year, year^2	1.036747	4	7.98	0.01	0.73
dieldrin	↔	%lipid, δ^{15}N, year	0.000855	5	0.00	0.31	0.18
		%lipid, δ^{15}N	0.000886	4	0.69	0.22	0.16
		%lipid, year^2	0.000893	4	1.34	0.16	0.16
		%lipid, δ^{15}N, year, year^2	0.000846	6	1.45	0.15	0.18
		%lipid	0.000929	3	2.41	0.09	0.13
ΣMirex	↓	%lipid, year	0.000694	4	0.00	0.44	0.29
		%lipid, δ^{15}N, year	0.000683	5	0.94	0.27	0.29
		%lipid, year, year^2	0.000692	6	2.03	0.16	0.28
Σ_{35}PCB	↓	%lipid, δ^{15}N, year^2	0.698911	6	0.00	0.52	0.83
		%lipid, year, year^2	0.719953	5	0.14	0.48	0.83
		%lipid, year	0.883343	4	14.85	0.00	0.79

[a] Chlorobenzenes (ΣCBz = 1,2,4,5-tetrachlorobenzene, 1,2,3,4-tetrachlorobenzene, pentachlorobenzene and hexachlorobenzene (HCB)), hexachlorocyclohexanes (ΣHCH = α-, β- and γ-hexachlorocyclohexane), octachlorostyrene (OCS), heptachlor epoxide (HE), chlordane-related compounds (ΣCHL = oxychlordane, *trans*-chlordane, *cis*-chlordane, *trans*-nonachlor and *cis*-nonachlor), DDT and its metabolites (ΣDDT = *p,p′*-DDE, *p,p′*-DDD and *p,p′*-DDT), dieldrin, ΣMirex = photomirex and mirex, and PCB congeners (Σ_{35}PCB).
[b] Trend direction was determined by backward stepwise regression analysis with year, % lipid and δ^{15}N as regressors. Non-significant ($p > 0.05$) time trends are indicated as "↔".

Table 3
Akaike's Information Criterion (ΔAICc, corrected for small sample sizes) values for representative models explaining variation in organochlorine concentration in eggs of black-legged kittiwakes breeding at Prince Leopold Island, NU, 1975–2013. Models compared linear and quadratic regressions to describe trends in organochlorine concentrations with year, year^2, %lipid and δ^{15}N as independent factors; model weights are presented (w_i^c) as is the R^2 from each GLM. We present only where the difference between model fit was small (ΔAICc <2), followed by the next best-fitted model.

Compound[a]	Trend[b]	Model	RSS	*K*	*ΔAICc*	w_i^c	R^2
ΣCBz	↓	year, year^2	0.005934	4	0.00	0.59	0.84
		%lipid, year, year^2	0.005761	5	1.94	0.22	0.82
		δ^{15}N, year, year^2	0.005934	5	2.86	0.14	0.82
ΣHCH	↑	δ^{15}N, year, year^2	0.000112	5	0.00	0.62	0.69
		δ^{15}N, %lipid, year, year^2	0.000110	6	2.54	0.17	0.67
α-HCH	↓	δ^{15}N, year, year^2	0.000007	5	0.00	0.53	0.66
		year, year^2	0.000008	4	1.28	0.28	0.58
		δ^{15}N, %lipid, year, year^2	0.000007	6	3.10	0.11	0.64
β-HCH	↑	δ^{15}N, year, year^2	0.000086	5	0.00	0.43	0.81
		year, year^2	0.000096	4	0.55	0.33	0.80
		%lipid, year, year^2	0.000093	5	2.43	0.13	0.80
OCS	↓	year, year^2	0.000001	4	0.00	0.48	0.75
		%lipid, year, year^2	0.000001	5	1.19	0.27	0.75
		δ^{15}N, year, year^2	0.000001	5	2.05	0.17	0.75
HE	↔	year, year^2	0.000177	4	0.00	0.49	0.20
		%lipid, year, year^2	0.000175	5	2.51	0.14	0.18
ΣCHL	↓	%lipid, δ^{15}N, year, year^2	0.002220	6	0.00	0.71	0.57
		δ^{15}N, year, year^2	0.002286	5	2.81	0.175	0.50
ΣDDT	↓	%lipid, year, year^2	0.091712	5	0.00	0.48	0.73
		year, year^2	0.103435	4	0.87	0.31	0.70
		%lipid, δ^{15}N, year, year^2	0.090208	6	2.59	0.13	0.72
Dieldrin	↓	year, year^2	0.000111	4	0.00	0.61	0.55
		δ^{15}N, year, year^2	0.000110	5	2.47	0.18	0.54
ΣMirex	↓	%lipid, year, year^2	0.000455	4	0.00	0.39	0.52
		year, year^2	0.000455	4	0.07	0.38	0.53
		δ^{15}N, year, year^2	0.000409	6	2.24	0.13	0.48
Σ_{35}PCB	↓	year, year^2	3.356436	4	0.00	0.41	0.72
		%lipid, year, year^2	3.060295	5	0.01	0.41	0.75
		%lipid, δ^{15}N, year, year^2	2.967379	6	2.14	0.25	0.75

[a] Chlorobenzenes (ΣCBz = 1,2,4,5-tetrachlorobenzene, 1,2,3,4-tetrachlorobenzene, pentachlorobenzene and hexachlorobenzene (HCB)), hexachlorocyclohexanes (ΣHCH = α-, β- and γ-hexachlorocyclohexane), octachlorostyrene (OCS), heptachlor epoxide (HE), chlordane-related compounds (ΣCHL = oxychlordane, *trans*-chlordane, *cis*-chlordane, *trans*-nonachlor and *cis*-nonachlor), DDT and its metabolites (ΣDDT = *p,p′*-DDE, *p,p′*-DDD and *p,p′*-DDT), dieldrin, ΣMirex = photomirex and mirex, and PCB congeners (Σ_{35}PCB).
[b] Trend direction was determined by backward stepwise regression analysis with year, % lipid and δ^{15}N as regressors. Non-significant ($p > 0.05$) time trends are indicated as "↔".

subsequently analyzed by backward stepwise regression with year, % lipid and δ^{15}N as regressors to ascertain trend direction over the sampling period for all data sets. Residuals from the regression analyses were tested for normality using the Shapiro-Wilks' W test and the concentration data ln-transformed if the residuals were found to violate the assumption of normality.

The tabulated data are presented as arithmetic means in concentration units of ng g^{-1} wet weight (ww) for the organochlorines and ‰ for δ^{15}N (Tables S1–S5). Concentration data presented in the figures were lipid-normalized to facilitate comparisons among species.

3. Results and discussion

3.1. Chemical profiles

The primary organochlorines found in eggs of all five species were Σ_{35}PCB, ΣDDT (mainly *p,p′*-DDE), ΣCBz (mainly HCB) and ΣCHL (mainly oxychlordane) (Figs. 1 and 2, Tables S1-S5) although proportions varied by species and year. Over all the years sampled, ΣPCB and ΣDDT together comprised 66–91% of total OCs in kittiwake eggs, 61–86% in fulmar eggs, 79–86% in glaucous gulls, but only 44–70% in murre eggs and 34–56% in guillemot eggs (Figs. 1 and 2). The fraction of Σ_{35}PCB decreased significantly in eggs of kittiwakes, fulmars and murres from 1975 to 2013–15, as did ΣDDT in the fulmar and murre eggs (Fig. 1, Table S11). These decreases were offset by increases in proportions of ΣCBz, ΣHCH (β-HCH), OCS, heptachlor epoxide, ΣCHL, dieldrin and ΣMirex (Fig. 1, Table S11).

A comparison among the five species for only the years in which eggs of all species were sampled (i.e. 1993, 1998, 2003/04, 2008, 2013) also showed a decrease in the Σ_{35}PCB fraction for all species except glaucous gulls, as well as decreases in ΣDDT fractions in eggs of all species (Fig. 2, Table S12). These decreases were offset by proportionate increases in ΣCBz (except in glaucous gulls), ΣHCH (β-HCH), OCS, heptachlor epoxide, ΣCHL, dieldrin and ΣMirex (Fig. 2, Table S12).

The OC profiles found among the species in this study were similar to those observed by Buckman et al. (2004) for the same species in the Northwater Polynya and by Borgå et al. (2001) for seabirds in the Barents Sea. Compositional profiles in murre and fulmar livers sampled from Prince Leopold Island in 2008 were also similar to those found in the murre and fulmar eggs from 2008 with ΣDDT > ΣPCB > ΣCBz > ΣCHL in the murres and ΣPCB > ΣDDT > ΣCHL > ΣCBz in the fulmars (Braune et al., 2014). Buckman et al. (2004) noted that the relative OC proportions followed a taxonomic family relationship, that is, greater

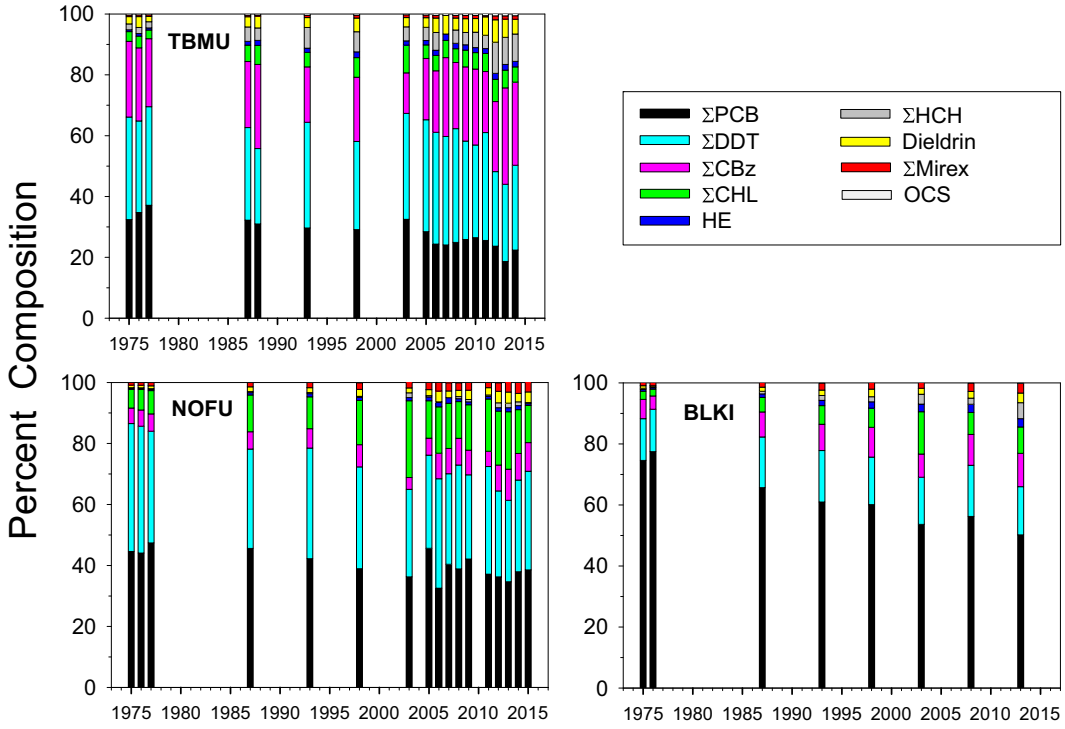

Fig. 1. Percent contributions of major organochlorine compounds/groups to total organochlorines (ΣOC) in eggs of black-legged kittiwakes (BLKI), northern fulmars (NOFU) and thick-billed murres (TBMU) from Prince Leopold Island, 1975–2015.

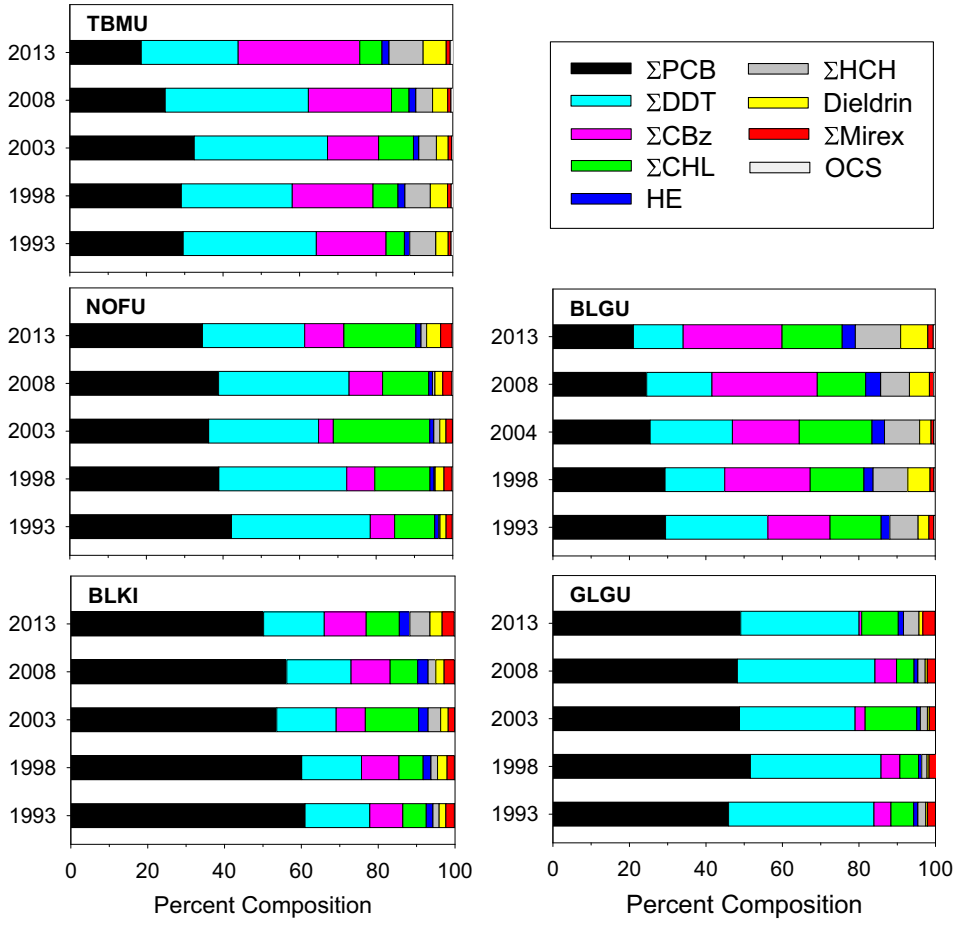

Fig. 2. Percent contributions of major organochlorine compounds/groups to total organochlorines (ΣOC) in eggs of black-legged kittiwakes (BLKI), northern fulmars (NOFU), thick-billed murres (TBMU), black guillemots (BLGU) and glaucous gulls (GLGU) from Prince Leopold Island for five comparative sampling years: 1993, 1998, 2003/2004, 2008, 2013.

proportions of ΣPCB and ΣDDT in larids (kittiwakes, gulls) and procellariids (fulmars) relative to the alcids (murres, guillemots), and greater relative amounts of ΣCHL, ΣCBz and ΣHCH in the alcids compared with the larids and procellariids. Differences in proportions of OCs among seabird species may be driven by differences in rates of change of compounds over time, biomagnification processes linked to trophic position, diet, metabolic capacities, as well as migratory habits and overwintering areas (Borgå et al., 2004; Fisk et al., 2001a).

ΣDDT was comprised almost entirely of *p,p′*-DDE which averaged >97% of ΣDDT in kittiwake, glaucous gull and guillemot eggs, 89–100% in murre eggs, and 86–94% in fulmar eggs across all years sampled (Tables S1-S5). In the murre and fulmar eggs, the relative proportions of *p,p′*-DDE in ΣDDT significantly increased ($r_s = 0.244$, $p = 0.029$ and $r_s = 0.469$, $p < 0.001$, respectively) as proportions of *p,p′*-DDD and *p,p′*-DDT decreased from 1975 to 2014/15 indicating decreased exposure to DDT over the study period and transformation of the parent compound to its major metabolite *p,p′*-DDE.

HCB averaged >80% of ΣCBz in all species. However, relative proportions of HCB in ΣCBz significantly decreased in kittiwake, murre and fulmar eggs ($r_s = -0.746$, $p < 0.001$; $r_s = -0.605$, $p < 0.001$; and $r_s = -0.707$, $p < 0.001$, respectively) from 1975 to 2013/14/15, as proportions of the tetra- and pentachlorobenzenes increased. Given that the primary use of HCB in agriculture was banned in the 1970s (Barber et al., 2005), the shift in the ΣCBz profile from mainly HCB to greater proportions of lower-chlorinated chlorobenzenes may reflect gradual dechlorination of HCB in the environment (Bailey et al., 2009; Barber et al., 2005).

ΣCHL was predominantly made up of oxychlordane for all species, averaging 81% in eggs of black-legged kittiwakes and northern fulmars, 73% in thick-billed murres, 89% in glaucous gulls, but only 40% in black guillemot eggs where *cis*- and *trans*-nonachlor averaged 35% and 22% of ΣCHL, respectively. As oxychlordane is the major metabolite of the chlordanes and nonachlors, it would appear that guillemots have a lower capacity to biotransform chlordane. This observation was also made by Fisk et al. (2001b) for guillemots in the Northwater Polynya relative to other seabird species and by Vorkamp et al. (2004) for guillemots in Greenland. Borgå et al. (2001) suggested that the higher contribution of oxychlordane to ΣCHL in kittiwakes and glaucous gulls compared with thick-billed murres and black guillemot may indicate a higher metabolic activity and excretion efficiency in gulls relative to murres and guillemots. The proportion of oxychlordane significantly decreased in kittiwake and fulmar eggs ($r_s = -0.372$, $p = 0.039$ and $r_s = -0.386$, $p < 0.001$, respectively) as proportions of *cis*-nonachlor significantly increased in kittiwake eggs ($r_s = 0.395$, $p = 0.028$) and *trans*-nonachlor significantly increased ($r_s = 0.406$, $p < 0.001$) in fulmar eggs from 1975 to 2013/15. These increases suggest possible continued exposure of those two species to chlordane components on their respective overwintering grounds at mid-latitudes, those being off the Newfoundland-Labrador Shelf in the case of the kittiwakes (Frederiksen et al., 2012), and across the North Atlantic in the case of the fulmars (Mallory et al., 2008, 2012).

ΣMirex was comprised of similar proportions of mirex and its photodegradation product, photomirex, which averaged 47–50% mirex and 50–54% photomirex in eggs of all species over years sampled except in guillemot eggs where proportions of photomirex averaged somewhat higher at 64% versus 36% for mirex. The proportional contribution of photomirex to ΣMirex increased in eggs of all species, significantly so in eggs of kittiwakes (1975–2013; $r_s = 0.415$, $p = 0.020$), fulmars (1975–2015; $r_s = 0.488$, $p < 0.001$) and guillemots (1993–2013; $r_s = 0.539$, $p = 0.020$) as the mirex fraction decreased, likely reflecting the photodegradation of mirex to photomirex in the environment (Gandhi et al., 2015).

The composition of ΣHCH differed among species. In eggs of murres and fulmars, α-HCH was the predominant isomer in the mid-1970s, but progressively decreased as β-HCH increased proportionately from <40% in the mid-1970s to >85% in recent years ($r_s = 0.921$, $p < 0.001$ and $r_s = 0.886$, $p < 0.001$, respectively). In guillemot eggs, a progressive shift from 57% α-HCH in 1993 to 91% β-HCH in 2013 was also observed. In kittiwake eggs, β-HCH was the predominant isomer over the entire period from 1975 to 2013 but also increased proportionately from 66 to 70% in the mid-1970s to >95% in 2008 and 2013 ($r_s = 0.842$, $p < 0.001$). Similarly, β-HCH also dominated the ΣHCH profile in glaucous gull eggs from 1993 to 2013, averaging 94%. γ-HCH was only intermittently detected in kittiwake, fulmar and murre eggs, and not at all in the guillemot and glaucous gull eggs (Tables S1–S5). Seabirds readily metabolize γ- and α-HCH although this biotransformation ability seems to vary among species with murres, guillemots and fulmars having a lower capacity to metabolize α-HCH than the kittiwakes and gulls (Borgå et al., 2001; Moisey et al., 2001). This pattern was reflected in the HCH profiles among species in this study. In contrast, β-HCH is recalcitrant and biomagnifies in the food web (Borgå et al., 2004; Moisey et al., 2001).

3.2. PCB homolog profiles

Major PCB congeners found in the eggs, that is, those congeners each averaging >5% of Σ_{35}PCB over the years sampled, included the following, in order of decreasing contribution: kittiwakes – CB-153, -138, -180, -118 and -99; fulmars – CB-153, -180, -138 and -118; murres – CB-153, -138, -118, -187, -99 and -180; guillemots – CB-153, -138, -118, -99, -187 and -180; glaucous gulls – CB-153, -138, -180 and -118. The penta-, hexa- and heptachlorobiphenyl homologs constituted the largest proportion of Σ_{35}PCB in all five species (Figs. 3 and 4). In the murres and guillemots, however, the lower chlorinated biphenyls (tri- to pentachlorobiphenyls) constituted a larger proportion of Σ_{35}PCB than in the other three species (Fig. 4). From 1975 to 2013–15, however, there seems to have been a significant increase in the fraction of hexachlorobiphenyls in the kittiwake, fulmar and murre eggs, as the proportions of the lower (except for the trichlorobiphenyls) and higher chlorinated biphenyls decreased (Table S13). A comparison among the five species for only the years in which eggs of all species were sampled (i.e. 1993, 1998, 2003/04, 2008, 2013) also showed a consistent increase in the hexachlorobiphenyl fraction as well as trichlorobiphenyls across species, offset by a consistent decrease in the hepta- and octachlorobiphenyls (Table S14).

Of the major PCB congeners found, i.e. CB-138, -153, -180, -118, -187 and -99, all are recalcitrant or not very readily metabolized (Borgå et al., 2005; Drouillard et al., 2007; Wania and Su, 2004) and CB-138, -153, -180, -99 and -187 have relatively high food web biomagnification factors (Borgå et al., 2004; Fisk et al., 2001a; Kelly et al., 2007). PCBs with an intermediate degree of chlorination have a higher Arctic Contamination Potential than the lesser or higher chlorinated congeners (Wania, 2006). Penta-, hexa- and hepta-chlorinated congeners were commercially produced in greater quantities than other congeners and have the highest bioaccumulation potential (McFarland and Clarke, 1989), and so it is not surprising that they dominated the PCB profiles. The tri- and tetrachlorinated congeners are more readily metabolized, while the higher chlorinated congeners are more tightly bound to particles and are, therefore, less bioavailable (Borgå et al., 2001; McFarland and Clarke, 1989; Wania, 2006). Further, there appears to be a tendency towards a lower maternal transfer of the more highly chlorinated (i.e. hepta- and octa-) congeners to the egg in some seabird species (Braune and Norstrom, 1989; Verreault et al., 2006). The difference in PCB profiles (i.e. greater proportion of lower chlorinated congeners) for murres and guillemots was also observed by Borgå et al. (2001) for seabirds in the Barents Sea and suggests that gulls and fulmars have a greater capacity to metabolize PCBs than alcids (Borgå et al., 2001, 2005).

3.3. Comparisons of organochlorine concentrations among species

Glaucous gull eggs had the highest concentrations of all organochlorines in all five years compared except for ΣCBz in 2013 where black

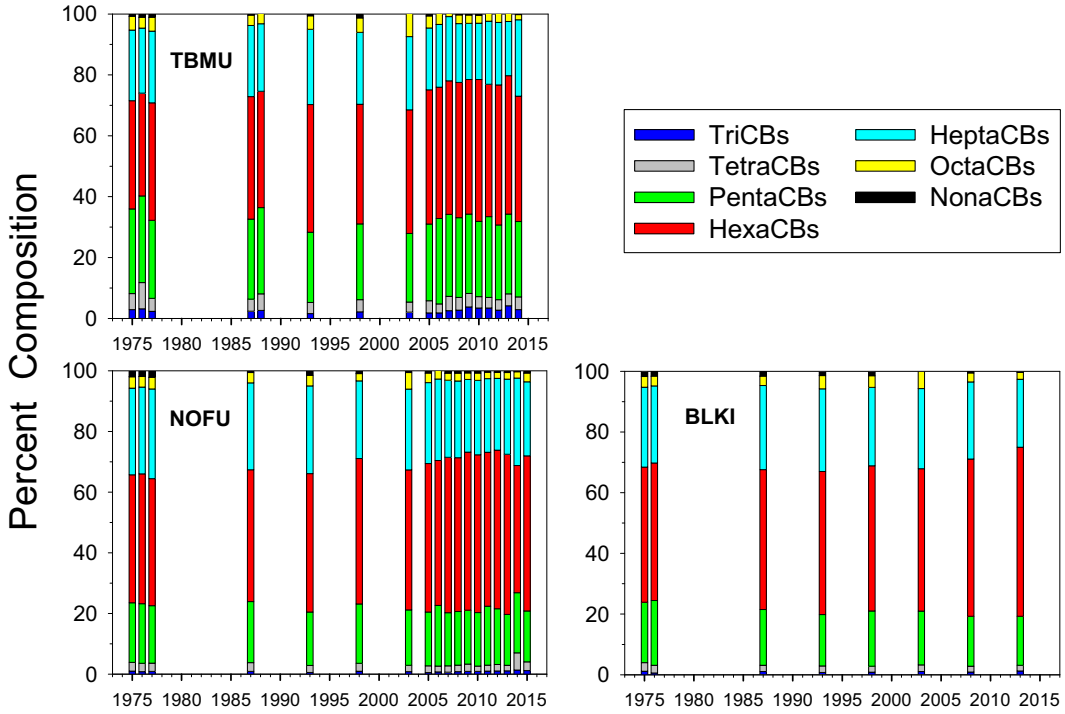

Fig. 3. Percent contributions of PCB homologs to Σ_{35}PCB in eggs of black-legged kittiwakes (BLKI), northern fulmars (NOFU) and thick-billed murres (TBMU) from Prince Leopold Island, 1975–2015.

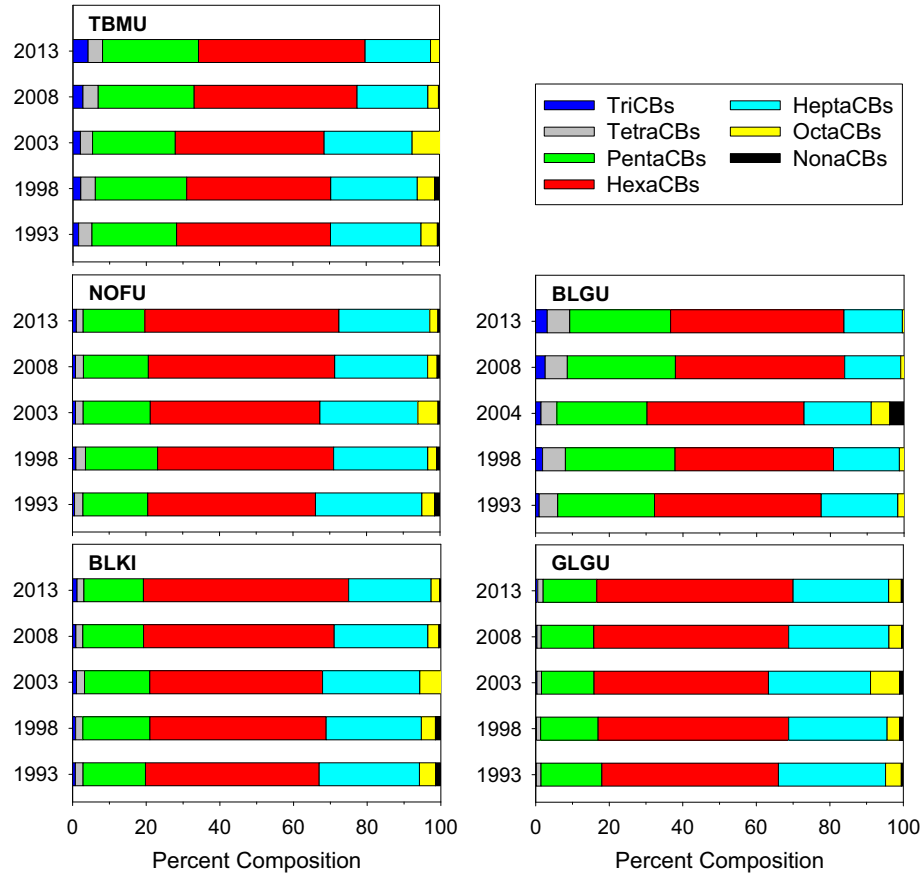

Fig. 4. Percent contributions of PCB homologs to Σ_{35}PCB in eggs of black-legged kittiwakes (BLKI), northern fulmars (NOFU), thick-billed murres (TBMU), black guillemots (BLGU) and glaucous gulls (GLGU) from Prince Leopold Island for five comparative sampling years: 1993, 1998, 2003/2004, 2008, 2013.

guillemot eggs had the highest ΣCBz concentration and glaucous gull eggs had the lowest mean ΣCBz concentration (Fig. 5). The difference was particularly marked for ΣDDT, Σ_{35}PCB and ΣOC for which concentrations were at least an order of magnitude greater in glaucous gull eggs than in the other four species. In 1993, fulmar eggs had two to three times greater ΣOC concentrations than eggs of kittiwakes, guillemots or murres, although the magnitude of these differences progressively diminished over time. Murre eggs generally had among the lowest concentrations of Σ_{35}PCB, ΣCHL and ΣOC (Fig. 5).

The δ^{15}N values for the glaucous gull eggs were relatively higher than in the other four species in 1993, 1998 and 2003/04 (Fig. 6), which aligned with their relatively greater organochlorine concentrations in those same years (Fig. 5), but the interspecific δ^{15}N patterns did not entirely explain the relative interspecific OC concentration patterns observed in any given year. For example, fulmar eggs had the lowest δ^{15}N values across all five years (Fig. 6) which did not necessarily align with their OC concentrations relative to the other species, particularly the kittiwakes, guillemots and murres. This same observation was also made by Buckman et al. (2004) for seabird species sampled from the Northwater Polynya in 1998. It was suggested that the discrepancies could be associated with factors such as occasional increased exposure to OCs via scavenging on marine mammals by species such as fulmars and glaucous gulls (Buckman et al., 2004; Hobson et al., 2002).

Glaucous gulls generally occupied among the highest trophic positions relative to the other seabird species in our study. In the 1998 study of the Northwater Polynya, seabirds were assigned to three trophic groups with the black-legged kittiwake, black guillemot, northern fulmar and thick-billed murre occupying intermediate positions and the glaucous gull occupying the highest trophic position (Hobson et al., 2002). The same relative trophic groupings were also observed by Hobson and Welch (1992) for those same five seabird species sampled from the Barrow Strait-Lancaster Sound marine food web during 1988–90.

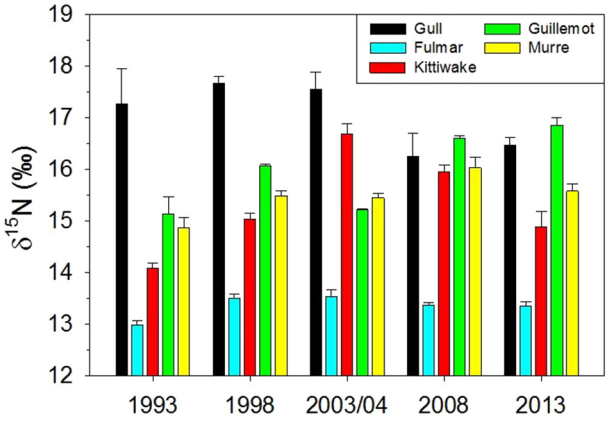

Fig. 6. Annual mean (±standard error) δ^{15}N (‰) values in eggs of major organochlorine compounds/groups in eggs of glaucous gulls, northern fulmars, black-legged kittiwakes, black guillemots and thick-billed murres from Prince Leopold Island for five comparative sampling years: 1993, 1998, 2003/2004, 2008, 2013. Modified from Braune et al. (2016).

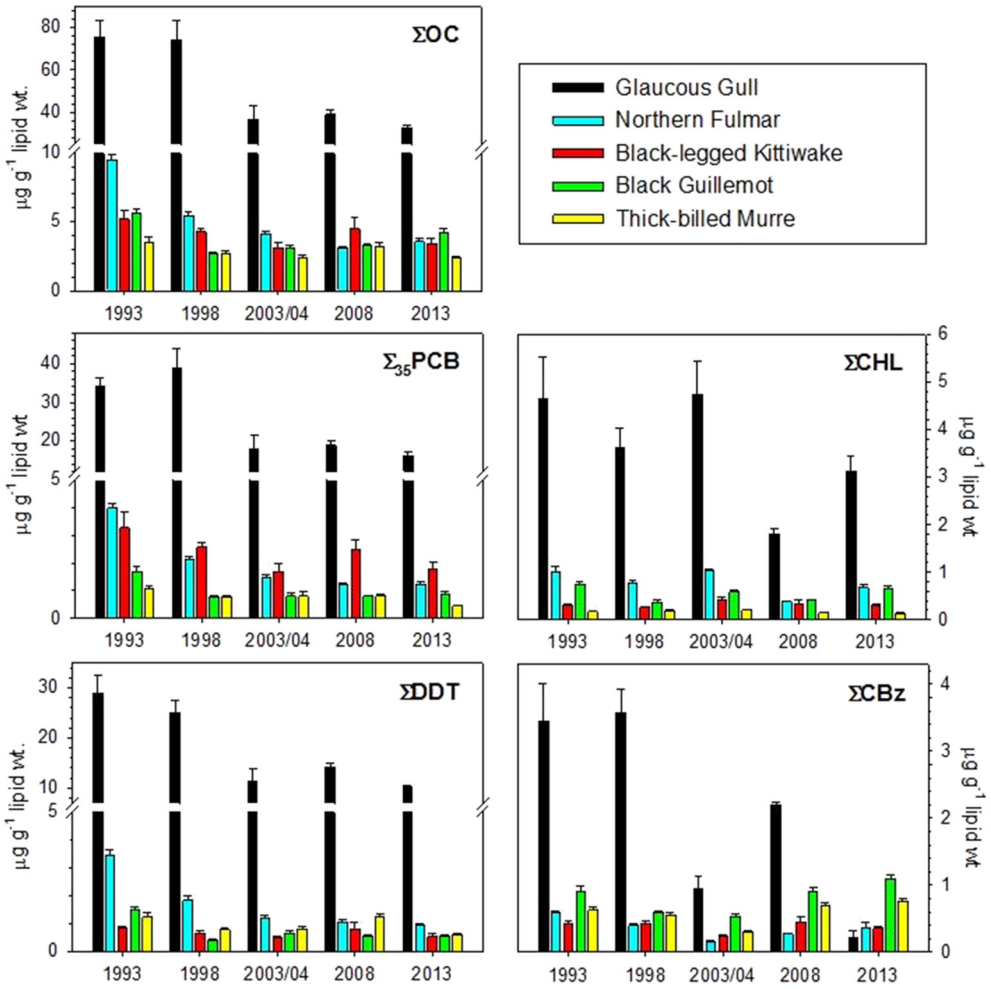

Fig. 5. Annual mean (±standard error) concentrations (μg g^{-1} lipid wt) of major organochlorine compounds/groups in eggs of glaucous gulls, northern fulmars, black-legged kittiwakes, black guillemots and thick-billed murres from Prince Leopold Island for five comparative sampling years: 1993, 1998, 2003/2004, 2008, 2013.

As observed in our study and a previous report of OCs in seabird eggs sampled from Prince Leopold Island in 1993 (Braune et al., 2002), glaucous gulls had higher concentrations of most organochlorines than other seabird species sampled from the Northwater Polynya in 1998 (Buckman et al., 2004) and the Barents Sea in 1995 (Borgå et al., 2001). Glaucous gulls have a broad diet although, as stated earlier, the gulls at Prince Leopold Island probably feed principally on marine prey during the breeding season (Matley et al., 2012; Nettleship et al., 1990). However, the glaucous gulls breeding on Prince Leopold Island probably accrue higher levels of OCs from two particular sources prior to egg-laying. First, Inuit have reported increasing numbers of large gulls in the spring that scavenge at Arctic community dumps and especially on marine mammal carcasses left near the community (Mallory et al., 2003; Mallory, unpublished data). Second, glaucous gulls feed on the eggs and nestlings of other seabirds (Nettleship et al., 1990), which may partially account for their higher trophic position and higher OC concentrations relative to the other seabird species. Some fulmars at Prince Leopold Island lay their eggs before glaucous gulls do (Gaston et al., 2005) and, therefore, it is likely that some adult gulls feed on fulmar eggs prior to laying their own eggs. Bustnes et al. (2000) also reported differences in OC concentration between two colonies of glaucous gulls in the Barents Sea, with the gulls that had a high proportion of seabird eggs in their diet having significantly higher OC concentrations in their blood compared with gulls feeding primarily on fish.

The relatively high PCB concentrations observed in the kittiwake eggs in our study may be attributed to the fact that they migrate to the eastern seaboard of North America to overwinter, placing them closer to historical sources of PCBs (Braune, 2007; Buckman et al., 2004). The consistently low ΣCHL concentrations seen in the thick-billed murre eggs (Fig. 5) may be related to the fact that murres appear to have a greater capacity to metabolize and eliminate chlordane relative to other species (Fisk et al., 2001b).

3.4. Temporal trends 1975–2015

Concentrations of most of the major organochlorine groups/compounds have either declined (e.g. Σ_{35}PCB, ΣDDT, ΣCBz, ΣCHL, OCS) or shown no consistent directional change (e.g. HE) since 1975 in eggs of murres, fulmars and kittiwakes (Tables 1–3, Figs. 7 and S2). For some compounds, such as the HCHs, as α-HCH was decreasing, β-HCH was increasing (Fig. S3), which resulted in no overall change over time for ΣHCH in the fulmar and murre eggs (Tables 1 and 2), or an overall increase in ΣHCH driven by the dramatic increase in β-HCH, as observed in the kittiwake eggs (Table 3, Fig. S3A). The best-fitting models describing either decreasing or increasing trends were all second-order polynomials with the exception of α-HCH, OCS and ΣMirex in the fulmar eggs which were linear decreases (Tables 1–3, Figs. S2 and S3). The best-fitting models (i.e. ΔAICc = 0.00) across compounds explained

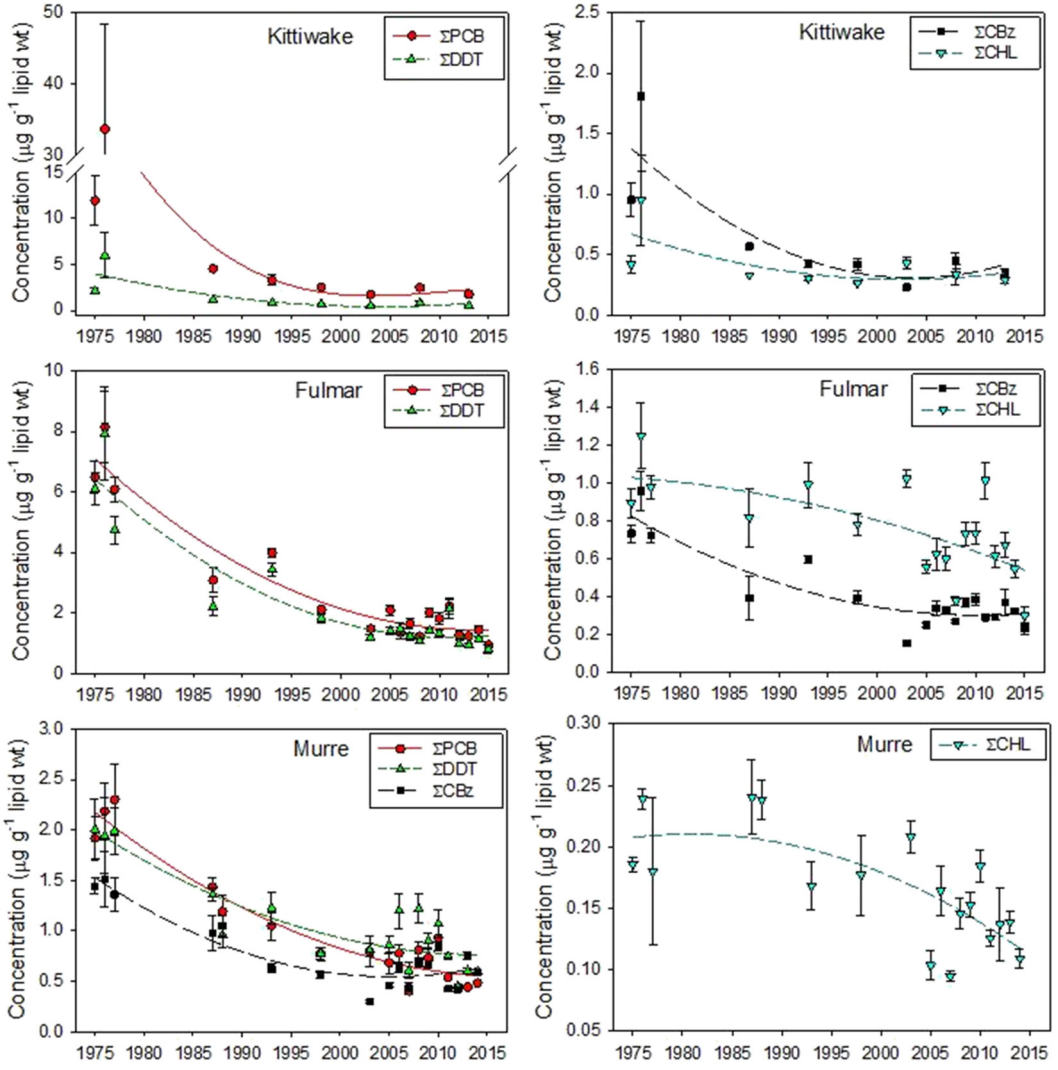

Fig. 7. Mean annual concentrations (μg g^{-1} lipid wt ± standard error) of Σ_{35}PCB, ΣDDT, ΣCBz and ΣCHL in eggs of black-legged kittiwakes, northern fulmars and thick-billed murres from Prince Leopold Island, Nunavut, Canada, 1975–2015.

24–73% of the variation for decreasing trends in the murre eggs, 29–83% in the fulmar eggs, and 52–84% in the kittiwake eggs (Tables 1–3). Equations for the best models matching GLM analyses are presented in Table S15.

Since 1975, most of the legacy POPs (e.g. PCBs, DDT) in the murre, fulmar and kittiwake eggs have been declining as has also been observed for other biota in North America (e.g. Champoux et al., 2015; de Solla et al., 2016; Mora et al., 2016) and elsewhere (e.g. Hammer et al., 2016; Helgason et al., 2012; Rigét et al., 2016). The trends for Σ_{35}PCB, ΣDDT and ΣCBz, in particular, have plateaued after the dramatic declines during the 1970s and 1980s resulting from the implementation of global and regional conventions which regulated or banned the use of most of the legacy POPs reported in our study (NCP, 2013; Wöhrnschimmel et al., 2016). The fact that most of the decreasing trends are best described by second-order polynomials indicates that the rate of uptake of legacy POPs is slowing or levelling off, suggesting that concentrations are reaching a steady state with environmental media and that trends are being less driven by primary sources and more by secondary sources such as environmental processing and degradation (Ma et al., 2011; NCP, 2013; Rigét et al., 2010; Wöhrnschimmel et al., 2016). Analyses of time-series datasets for circumpolar biota found that downward trends constituted the majority of statistically significant trends of the legacy POPs which had been subject to bans or regulations for 20–30 years, in particular, DDTs, HCHs, PCBs and chlordanes (AMAP, 2016). The lack of a trend for HE in eggs of all three seabird species in our study is consistent with the findings for other circumpolar datasets for biota and it has been suggested that this reflects the long half-life of HE in the environment resulting in a slow and gradually diminishing influence of re-emissions from past deposition (AMAP, 2016). Although we observed decreasing trends of ΣCBz in the eggs of all three seabird species since 1975, increasing trends have been seen in other Arctic biota suggesting that emissions of HCB are still occurring, likely as a by-product from the manufacture of chlorinated compounds, or as an impurity in other pesticides, or from industrial combustion sources (AMAP, 2016).

Mirex is an extremely volatile and persistent compound (Gandhi et al., 2015). Kittiwake eggs showed the greatest decline in ΣMirex concentrations from 1975 to 2013 (Fig. S2) and none of the five species showed a trend from 1993 to 2013 (Tables S6–S10), which is consistent with the observation that the major declines in Arctic biota occurred before 2000 (AMAP, 2016). Likewise, major declines of OCS concentrations occurred in eggs of all three species since 1975, consistent with trends in other Arctic biota (AMAP, 2016).

The lack of a temporal trend for ΣHCH in the seabird eggs except for the kittiwakes (see Tables 1-3 and Tables S6-S10) reflects the opposing trends of two of the HCH isomers. Consistent with primarily decreasing trends of α-HCH seen in other Arctic biota (AMAP, 2016), concentrations of α-HCH also decreased in eggs of all five seabird species, either from 1975 onwards (Tables 1–3, Fig. S3) or from 1993 to 2013 (Tables S6–S10). The decline in α-HCH was offset by increasing concentrations of β-HCH in the seabird eggs (Tables 1–3 and Tables S6–S10, Fig. S3). The increasing concentrations of β-HCH are consistent with the recalcitrant nature of this isomer as reflected by the increasing or major proportions of β-HCH found in the eggs of all five species (see section 3.1), as well as the fact that β-HCH biomagnifies in the food web, and seabirds appear to readily metabolize the γ and α isomers (Borgå et al., 2004; Hop et al., 2002; Moisey et al., 2001). There seems to be no consistent pattern to HCH trends among Arctic biota (AMAP, 2016), likely due to the variable biotransformation capacities among species (Moisey et al., 2001) compounded by the spatial differences in the presence of HCHs in Arctic Ocean surface waters; i.e. highest concentrations in the Beaufort Sea and Canadian Archipelago, lowest concentrations in the Barents Sea and eastern Arctic Ocean (Macdonald et al., 2000; NCP, 2013). These spatial differences can be attributed to the different physical-chemical properties of the HCH isomers leading to different transport pathways (α-HCH being transported by air and β-HCH primarily following a slower oceanic route) to the Arctic from their major source regions in southeast Asia (Li and Macdonald, 2005; NCP, 2013).

There is the suggestion of possible increasing concentrations of some compounds (e.g. dieldrin, OCS, HE) in kittiwakes eggs, in particular, in recent years (Fig. S2). This may be the result of remobilization and re-emission of POPs related to climate change (Kallenborn et al., 2012; Ma et al., 2011), or may simply reflect inter-annual variation in the data.

3.5. Comparison of temporal trends among five species

With fewer time points (i.e. five) over a shorter time frame (1993–2013), the temporal trends compared among the five species were less defined, although concentrations of Σ_{35}PCB and ΣDDT were still declining in eggs of all species except for ΣDDT in kittiwake eggs (Tables S6–S10, Fig. 8). Equations for the best models matching GLM analyses are presented in Table S16. Declines in the glaucous gull eggs were most pronounced, likely associated with glaucous gull diet and trophic status. It is clear that most of the declines occurred during the 1970s to 1990s followed by little change during the 2000s (Fig. 7) although the reverse appears to hold true for changes in the HCH concentrations where α-HCH continued to decline while β-HCH concentrations increased after 2000 (Fig. S3). The declines in α-HCH after 2000 are consistent with decreases observed in other Arctic biota (AMAP, 2016).

Aside from β-HCH, the major OC compounds either declined or showed no trend between 1993 and 2013. In fact, some compounds (e.g. ΣCBz) appeared to show an increase in recent years in the murre and guillemot eggs (Fig. 8) which supports evidence that emissions of HCB are still occurring (AMAP, 2016). Further, the greater relative amounts of ΣCBz found in the alcids compared with the larids and procellariids (Fig. 2) suggests that the murres and guillemots have a lower capacity to metabolize HCB.

4. Conclusions

Over four decades, our monitoring of POPs in eggs of seabird species suggests that most of the legacy OCs declined in the Arctic environment from the 1970s through the 1990s, and then has plateaued during the 2000s, albeit with clear differences among species and compounds. Although the interaction of the various physical-chemical, ecological and physiological factors driving the temporal trends of legacy organochlorines in seabird eggs is complex, trends in seabird eggs suggest that the environment does respond to reductions in contaminant emissions and that seabirds are good monitors of changing emission patterns of environmental pollutants. However, these trends may be altered by climate change as it affects the biogeochemical processes of environmental contaminants entering the Arctic as well as processes remobilizing earlier depositions (Hansen et al., 2015; Kallenborn et al., 2012; Ma et al., 2011; Macdonald et al., 2005; Noyes et al., 2009). Changes in food web structures may also occur which, in turn, can affect the biomagnification pathways of POPs (Braune et al., 2015b; Macdonald et al., 2005; McKinney et al., 2015; Nadal et al., 2015; Noyes et al., 2009). Finally, recent studies have found negative, sub-lethal effects of POPs on Arctic seabirds at environmentally-relevant concentrations (e.g. Blévin et al., 2016; Goutte et al., 2015; Letcher et al., 2010; Melnes et al., 2017; Tartu et al., 2014; Verreault et al., 2013). Therefore, it is important to continue to monitor these legacy contaminants in order to ascertain how environmental changes will affect the temporal trends observed to date, and to what extent there may be long-term effects on populations of Arctic seabirds.

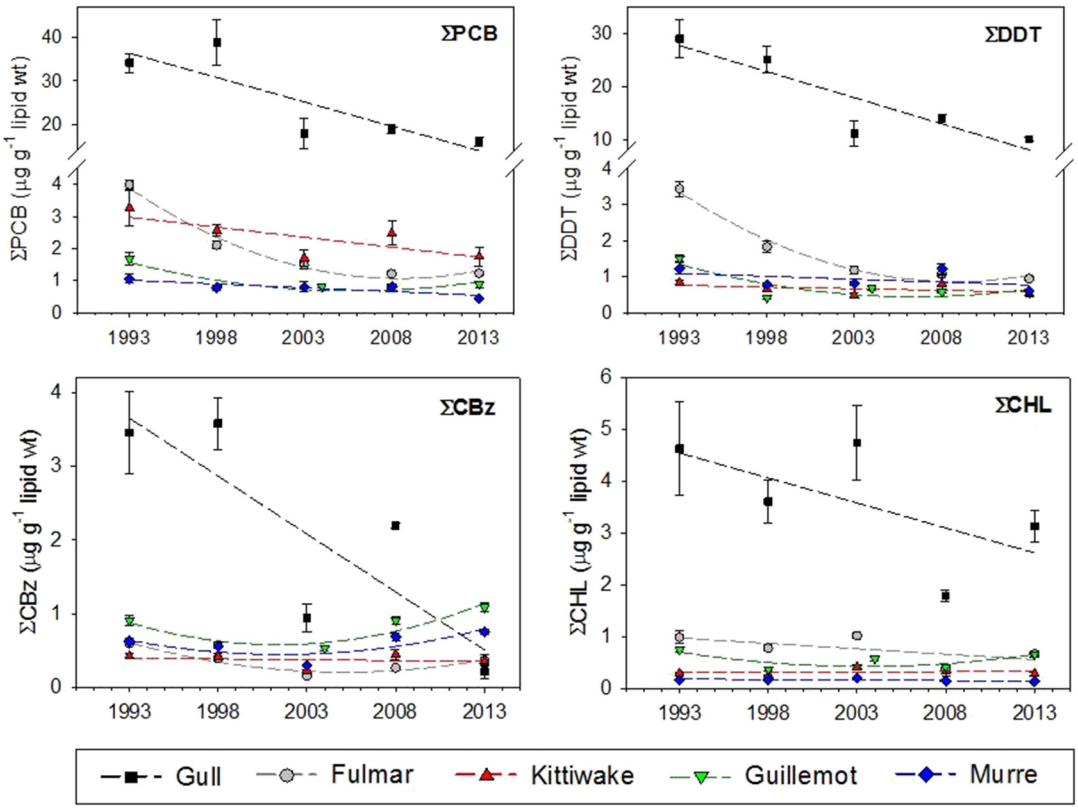

Fig. 8. Mean annual concentrations (µg g^{-1} lipid wt ± standard error) of Σ_{35}PCB, ΣDDT, ΣCBz and ΣCHL in eggs of black-legged kittiwakes, northern fulmars, thick-billed murres, black guillemots and glaucous gulls from Prince Leopold Island, Nunavut, Canada, 1993–2013.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2018.07.291.

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